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Anisotropic magnetoresistance of $La_{0.7}Ca_{0.3}MnO_3$ thin film biepitaxial step junctions

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The angular dependence of magnetoresistance (MR) of the $La_{0.7}Ca_{0.3}MnO_3$ thin film biepitaxial step junction (BSJ) shows a simple $\sin^2(\theta)$ dependence in the in-plane high-field magnetoresistance, with θ being the angle between the applied field and current. This behavior is similar to the spin-orbit coupling-induced anisotropic magnetoresistance (AMR) commonly observed in transition ferromagnetic metals, except for two salient features. First, the maximum MR in the present case occurs at an oblique angle between the applied field (H) and electric current (I), while it is usually observed to occur when $H \| I$. Second, the AMR in the plane perpendicular to the film surface displays a remarkable value ($\Delta\rho/\rho \sim 8\%$), which is about an order of magnitude larger than that of the in-plane AMR. Such a large AMR cannot be solely explained by spin-orbit coupling effect. We suggest instead that the metallic and ferromagnetic inhomogeneous granules existing in the BSJ region might have acted as the source of spin-polarized scattering giving rise to the enhanced AMR when the colossal magnetoresistance was measured across the biepitaxial step boundaries. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2390545]

I. INTRODUCTION

The grain-boundary magnetoresistance (GBMR) have been drawing much attention since the discovery of colossal magnetoresistance (CMR) in granular manganites.[1,2] From technological point of view, it is always attractive if practical MR ratio can be routinely obtained without resorting to the more complicated structure such as multilayer junctions commonly used in giant MR (GMR) or tunneling MR (TMR) devices.[3] For this application purpose, the single layer artificial GBMR devices are of potential importance. Fundamentally, however, the understanding of the basic mechanisms giving rise to the observed fascinating GBMR remains unsettled.[4–10] For instance, in most artificial GB junctions where the current-voltage characteristics (IVCs) were nonlinear,[6,8,11] the low-field MR (defined as MR measured at fields below the magnetization saturation, which is typically 0.1 T for relevant manganites) of the GB junctions has been attributed to the spin-polarized tunneling via insulating GB regions between ferromagnetic grains, whereas, for those displayed Ohmic IVCs, the spin-polarized scattering between ferromagnetic grains has been proposed to account for the large low-field MR.[1,9,10] Moreover, as the field increases beyond 0.1 T, the MR continues to decrease with applied field. This high-field MR has been consistently observed in most types of the GB junctions and is almost independent of temperature below the ferromagnetic transition temperature. Both the spin-polarized tunneling and spin-polarized scattering models predict that the resistance should remain unchanged once the magnetization of ferromagnetic grains saturates. Thus, both are inadequate to explain the appearance of the high-field MR.

On the other hand, the anisotropic magnetoresistance (AMR) of GBs has also been extensively discussed[11–15] owing to the importance of AMR effect on the operation of conventional magnetoresistive read heads used in magnetic storage devices. Fundamentally, the high-field AMR effect is a well known inherent property of ferromagnets originated from the local spin-orbit scattering of conduction electrons.[16,17] In ferromagnetic metals, such as iron, cobalt, and nickel, the AMR is directly related to the magnetization and leads to a direct dependence on the square of the magnetization component transverse to the current direction, provided that the applied field is much larger than the coercive field. The angular dependent MR can be described fairly well by the relation $R(\theta)/R(0) = 1 + A \times \sin^2(\theta)$, where θ is the angle between the magnetization and the electric current, and A is the amplitude of AMR. Thus, in a saturated ferromagnet, the resistivity should have an angular dependence on the direction of current flow that is temperature and field independent. Furthermore, it should vanish as the temperature is increased to T_C and above. Based on this concept, Ziese *et al.*[6,18] have investigated the AMR in various types of the GBMR junctions and suggested that electron tunneling between the highly spin-polarized ferromagnetic grains through magnetic manganese atoms in the insulating GB region dominates the high-field MR. However, as mentioned above, tunneling-related mechanisms seem to be inconsistent with

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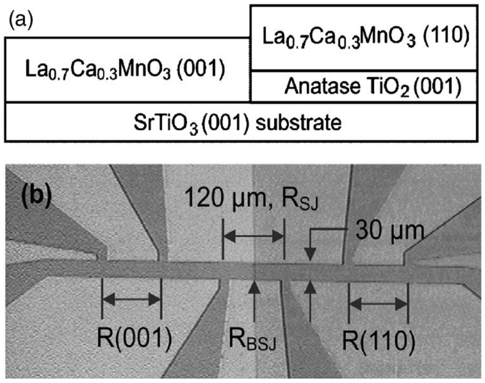

FIG. 1. (a) The cross-sectional schematics of the biepitaxial sample. The $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) film grown on the $SrTiO_3$ substrate exhibits (001) orientation, whereas the LCMO film grown on the TiO_2 buffer layer is (110) oriented. (b) The top-view microscopic image of the patterned biepitaxial sample shown in (a). The $R(001)$, $R(110)$, and R_{SJ} represent the resistances of the respective segments.

the Ohmic IVCs found in some cases,[4,9,10] where it is indicative that the extent of magnetic order in the GB regions may have played a role in high-field MR. It is thus desirable to conduct investigations that link these seemingly related phenomena to provide a deeper insight into these materials of emerging importance.

In this paper, we report results obtained from measuring the angular dependence of MR on the $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) biepitaxial step junctions (BSJs) obtained by depositing LCMO film onto $SrTiO_3(100)$ substrate partially buffered with a layer of anatase $TiO_2(001)$.[10] The BSJ shows an unusually large AMR ratio and the maximum MR occurs at an angle of 50° between the electric current and applied field, indicating that the boundary region, when magnetized at relatively high fields, may still be rather inhomogeneous. This is consistent with the conjectures of attributing the high-field MR to polarized spin scattering through the inhomogeneous metallic and ferromagnetic granules existent within the boundary regions.[10] It is noted that recent microstructure analyses on manganite films by Soh *et al.*[19,20] also gave direct evidences of strain-induced magnetic inhomogeneities in the vicinity of grain boundaries.

II. EXPERIMENT

The cross sectional structure of the BSJ sample is schematically illustrated in Fig. 1(a). Detailed description of the fabrication process of the BSJs has been reported previously.[10] Briefly, the LCMO films grown on the region of bared $SrTiO_3$ substrate and that covered by the TiO_2 buffer layer are (001) and (110) oriented, respectively. The lattice constants of LCMO (assuming cubic) calculated from both (001) and (110) are 0.3859 and 0.3856 nm, respectively. Since the difference is only about 0.07%, we presume that the distortion is not significant and the transport results to be discussed below may not be much relevant to effects arising from lattice distortion. The samples were patterned into bridge using wet etching method to produce a BSJ. Figure 1(b) shows the microscopic image of the patterned biepitaxial sample. The BSJ indicated by the arrow, thus, forms a misoriented GB junction across the boundary separating (001) and (110) regions. The contact pads at each side of the BSJ are used to perform the four-probe resistance measurements of the patterned bridge with and without crossing the BSJ. In this way, the BSJ resistance R_{BSJ} can be obtained by subtracting the resistance of the segment in (001) region, $R(001)$, and that in (110) region, $R(110)$, from the resistance of the whole segment R_{SJ}. The magnetotransport measurements were carried out in a Quantum Design PPMS® system with the angular resolution being better than 1°.

Previously, we have demonstrated some important features of GBMR exhibited by this type of BSJ bridges.[10] They exhibit prominent low-field MR (LFMR) behaviors that are typical to most of other types of GB junctions, namely the resistance peaks at the coercive field (H_c) of the magnetization hysteresis loop with a MR ratio of about 20%. The field-dependent MR change normalized to resistance at H_c is also found to be proportional to the square of global magnetization, suggesting that either magnetic inhomogeneity-induced scattering[9] or intergrain spin-polarized tunneling[2] is responsible for the results. However, the fact that the high-field MR (HFMR) deviates significantly from expected linear dependence in H (Refs. 5 and 10) and the linear current-voltage characteristic[9,10] has led us to propose that the magnetotransport in this artificial structure may be dominated by polarized spin scattering across the boundary region, instead of attributing it to various types of tunneling mechanisms.[4–8]

III. RESULTS AND DISCUSSION

Figure 2(a) shows the typical temperature-dependent magnetization for the entire biepitaxial film prior to BSJ bridge patterning. There appears to be only one Curie temperature around 260 K for both the (001)- and (110)-LCMO films. The result indicates both the quality and homogeneity of the film. Figure 2(b) displays the M-H curve of the film at 5 K, showing that 90% of magnetization moment saturates at about 1000 Oe. The typical hysteresis loop further indicates that both LCMO(110) and LCMO(001) have the same coercive field $H_c \approx 250$ Oe. Although the above observations may merely reflect the isotropic nature of LCMO manganite, they, nevertheless, imply that the effects to be discussed below should be predominately due to the step junction.

Figure 3 shows the angular-dependent MRs of the BSJ at 75 and 150 K with a field of 3 T applied in the plane perpendicular to the film surface. The applied fields used in the measurements were sufficiently larger than the coercive field so that the saturated magnetization is presumed to align with the field. The zero-degree angle ($\theta=0$) was defined as the angle when the applied field was perpendicular to the current flow direction. Because the angular configuration is different from the usual equation $[R(\theta)/R(0)=1+A\times\sin^2(\theta)]$ used for describing the spin-orbit interaction-induced AMR, we add a constant θ_c into the $\sin^2(\theta)$ term and replace $R(0)$ with the minimum resistance R_{min}. The AMR equation, thus, becomes $R(\theta)/R_{min}=1+A\times\sin^2(\theta-\theta_c)$. The circles in the figures are the experimental data, whereas the solid lines are the fittings using the above equation with A and θ_c being the only adjusting parameters. As is evident from Fig. 3, the fits to the data are fairly satisfactory, suggesting that it might have

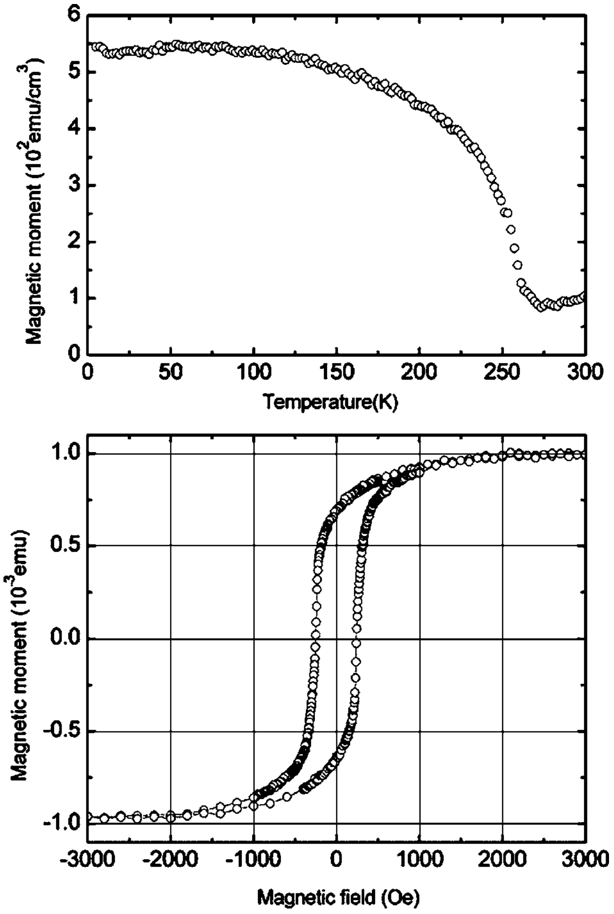

FIG. 2. (a) The temperature-dependent magnetization, $M(T)$, and (b) the M-H curve measured at 10 K of the biepitaxial LCMO films. The results indicate that both (001)- and (110)-LCMO have the same Curie temperature and coercive field without a noticeable impurity phase.

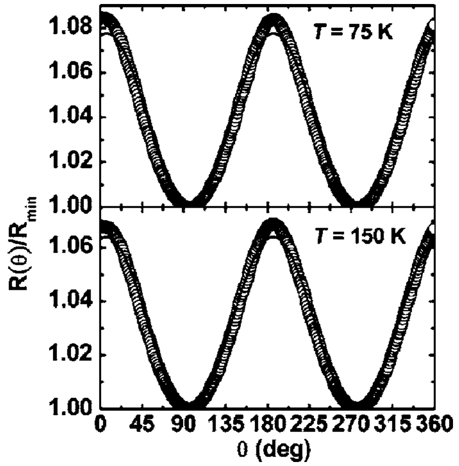

FIG. 3. The angular-dependent MR for the BSJ at 75 and 150 K with the applied field of 3 T. The field was applied in the plane perpendicular to the film surface. The circles are the experimental data, whereas the solid lines are the AMR fitting result.

originated from the same spin-orbit interaction prevailing in conventional ferromagnetic materials. Nonetheless, we note that the AMR ratio ($\sim 8\%$) is remarkably large and appears to be increasing at lower temperatures. The enhancement of AMR with decreasing temperature is a common feature in the granular manganite films.[12] Since the applied magnetic field was well above the saturation field, this large AMR ratio cannot be caused by the demagnetization effect. On the other hand, Ziese and Sena[21] have estimated the spin-orbit interaction-induced AMR and concluded that in $La_{0.7}Ca_{0.3}MnO_3$ films it should be less than 1%, which apparently cannot account for the large AMR effect observed in the current BSJ either. Since the high-field AMR is intimately related to the magnetization and the current direction, the current observation implies that significant magnetic anisotropy might be existent in the BSJ region. This could be due to the existence of preferential magnetization orientations.[9] In such scenario, the present results indicate that it is more difficult to align the magnetization in the BSJ along the orientation perpendicular to the film plane than that in the plane. In order to further elaborate the possible underlying mechanism, it is instructive to briefly revisit some of the previous experimental observations on these films.

As has been described previously,[10] the magnetization of the biepitaxial sample saturates at a field of about 0.1 T, which is also the field used to separate the low-field and high-field regimes in discussing the MR of the BSJ. The ferromagnetic transition of the LCMO (001) and LCMO (110) films, which serve as the two electrodes of the present BSJ, occurs at the same temperature (about 260 K) (Fig. 2), and below this temperature, the resistance of the entire BSJ starts to exhibit a sharp decrease in the low-field regime.[10] This resistance drop is indicative of switching of magnetic domains in the applied field. The low-field MR of the BSJ increases with decreasing temperature and reaches about 20%, a value comparable with many other types of artificial GB junctions.[5,6]

It is apparent that direct access to the information at the length scale of the GB regions is essential for understanding GBMR. However, a direct magnetic measurement of the GBs has not been achieved so far. Although the coexistence of insulatinglike and metalliclike phases at nanometer scale in a $La_{0.67}Ca_{0.33}MnO_3$ film have been evidently demonstrated[22] by scanning tunneling microscopy, it is very difficult to locate the BSJ region exactly. As a result, it is not clear how phase separation in the vicinity of GB accounts for the complexity of the resistance behaviors in manganites. The study of AMR, however, is directly associated with the BSJ region, and thus, may serve as an alternative probe for extracting the magnetic properties within the step junction.

The results of angular-dependent MR at low temperature (10 K) with various applied fields are illustrated in Fig. 4. At this temperature, a maximum AMR is expected in the granular films. As can be seen in the figure, the AMR can be well fitted by $\sin^2(\theta)$ at 1 T with slight deviation at high fields. Based on the assumption of the isotropic magnetization in the film plane, this AMR should arise from the same mechanism as in ferromagnetic metals. Indeed, the anisotropy magnitude is comparable to that expected from the spin-orbit scattering effect. However, it is striking to note that the angle of the MR maximum, i.e., the minimum in $R(\theta)/R_{\min}$, dramatically shifted to the lower angle. The fitted θ_c values are 55.9°, 55.5°, and 52.2° for the applied fields of 1, 3, and 8 T,

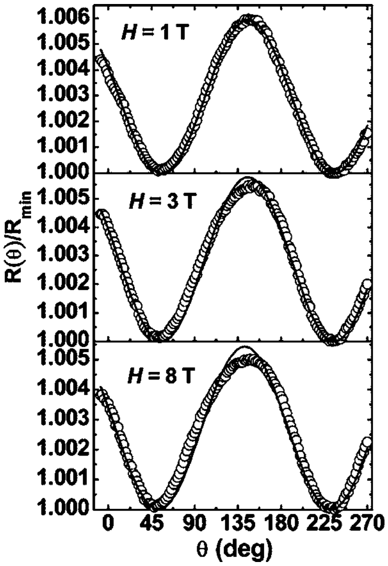

FIG. 4. The angular-dependent MR for the BSJ at 10 K with the various applied fields. The field was applied in the plane of the film surface. Angle zero is defined as the field perpendicular to the electric current. The circles are the experimental data, whereas the solid lines are the AMR fitting results.

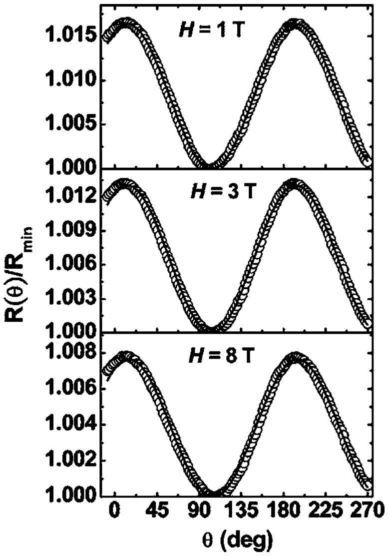

FIG. 5. The angular-dependent MR for the LCMO (110)+LCMO (001) film BSJ at 10 K with various applied fields. The field was applied in the plane of the film surface. Angle zero is defined as the field perpendicular to the electric current. The circles are the experimental data, whereas the solid lines are the AMR fitting results.

respectively. To further identify this peculiar behavior, we calculate the angular-dependent MRs of the LCMO (110) plus LCMO (001) films in the same plane.

As discussed above, the BSJ resistance is extracted by using the two pairs of measuring contact pads nearest to the BSJ. Similarly, the sum of the segment resistances belonging to the LCMO (001) and (110) films, which are indicated as $R(001)$ and $R(110)$ as shown in Fig. 1 can also be identified separately. This resistance sum, however, mostly reflects the angular-dependent MR of the LCMO (110) film because the resistance of the LCMO (110) film is more than one order of magnitude larger than that of the LCMO (001) film. This angular-dependent MR of the resistance sum is illustrated in Fig. 4. Although the experimental data can be fitted very well by $\sin^2(\theta)$, the fitted parameter θ_c has the value of about 104° and is independent of magnetic field. The 14° difference accounting for the angle between the magnetization and applied field direction, though may be due to the experimental misalignment, is more likely an intrinsic contribution from the LCMO (110) film. Nonetheless, the angular difference of about 50° between the resistance minima in the BSJ and LCMO clearly needs further discussion.

Recently, Blamire *et al.*[8] has found a similar effect in $La_{0.7}Ca_{0.3}MnO_3$ bicrystal junctions, in that the maximum MR occurs when the applied field is directed at an angle of 22° with respect to the GB. They attributed this deviation to the magnetocrystalline anisotropy. However, we noted that this anisotropy scenario probably could not be applied to the BSJ results shown above because the AMR was obtained with a very large applied field and the coercive fields of the LCMO (001) and (110) films in our BSJ sample are the same. Since the high-field AMR effect is mainly associated with the magnetization and transport current in ferromagnetic metals, the resistance minimum occurring at an essentially arbitrary angle may have resulted from magnetization inhomogeneity. That is, the magnetization in the BSJ region is not as easy to align with the applied field as that in the epitaxial regions. This magnetization inhomogeneity is inherent to the BSJ region because within it the crystal axis rotates from (001) to (110) orientation, and hence, may accompany a large strain field. As has been pointed out by Belevtsev *et al.*,[11] such a large strain field may lead to significant anisotropic properties in $La_{1-x}Ca_xMnO_3$ films. As can be seen in the present AMR results, the BSJ exhibits the feature of AMR, thus, it is suggestive that there could be a certain degree of magnetic order existent in the BSJ region. In addition, the magnitude of the AMR monotonically decreases with increasing magnetic field up to 8 T (Figs. 4 and 5). This is consistent with the monotonic decrease of the high-field resistance of the BSJ.[10] The fact that both the angular dependence of the large AMR effect and the high-field MR of the present BSJ can be consistently explained by the magnetic inhomogeneity in the GB region, further indicates that the magnetotransport properties of various artificial manganite junctions are dominated by the polarized spin scattering rather than tunneling mechanisms.

In summary, we have measured the AMR of the BSJ and found that the MR of the BSJ reaches maximum at an angle of 50° between the applied field and the electric current. The magnitudes of AMR, however, suggest that the conventional spin-orbit coupling effect, though may explain the in-plane anisotropy, is inadequate to account for the perpendicular anisotropy. The fact that the BSJ is metallic and displays a certain degree of magnetic order indicates that the AMR arises primarily from the inhomogeneity induced by the tremendous strain existent in the step region. Within this scenario, the MR of the BSJ can be consistently described by the spin-polarized scattering mechanism previously conjectured.

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